COMB-LOCKED CAVITY-ASSISTED DOUBLE RESONANCE (COCA-DR) SPECTROSCOPY OF MOLECULES WITH kHz ACCURACY

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Double resonance (DR) spectroscopy has been frequently applied in state-selective excitation and to reach energy levels forbidden to single-photon transitions. Due to the low cross section of two-photon transitions, usually high-power pulsed lasers are needed, which prevent high-precision measurements. Here we present a newly developed comb-locked cavity-assisted double resonance (COCA-DR) spectroscopy technique by simultaneously locking two diode lasers (1.60 μm and 1.67 μm) to a high-finesse cavity and an optical frequency comb. Doppler-free optical-optical DR transitions to the highly-excited (60025) ($v_{CO} = 8$) "dark" state of the CO_2 molecule using the (30013) vibrational state as the intermediate. By selecting molecules with different longitudinal speeds, we confirmed a frequency accuracy of 3 kHz of the measured transitions. Rotational energies of the (60025) state and the transition dipole of the (60025)-(30013) band were determined for the first time, and they were compared with the calculated values based on effective operators. The measurements demonstrate the possibility to determine energies of highly excited states of molecules with unprecedented precision.